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AN EXACT ONE-DIMENSIONAL SOLUTION TO THE PROBLEM OF CHLOROPHYLL--ETC(U)

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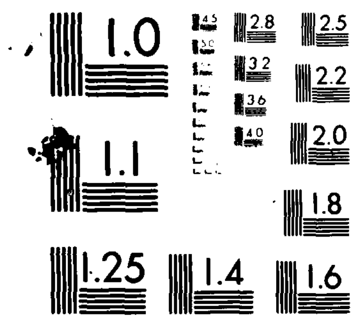

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AN EXACT ONE-DIMENSIONAL SOLUTION TO THE PROBLEM
OF CHLOROPHYLL FLUORESCENCE FROM THE OCEAN

by

George W. Kattawar and John C. Vastano

Report No. 16

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We have obtained an exact, one-dimensional solution to the equations of radiative transfer for chloropyll fluorescence from an infinitely deep ocean with a uniform distribution of fluorescing bodies. We have also found that the general solution could be reduced to a very simple expression for the case where absorption dominated over scattering. This allowed us to establish an upper bound on the difference in reflectivities for a very high chlorophyll concentration.

I. Introduction

The diffuse reflectance peaks occuring at ≈685 nm which were reported by Morel and Prieur[1] over water bodies bearing phytoplankton have been the subject of speculation over the past few years. Enhanced scattering from chlorophyll bearing cells has been observed for many years[2,3,4]. Even differential scattering of circularly polarized light by chloroplasts has been observed[5]. There seems to be little doubt that anomolous dispersion provides a suitable model for explaining the enhanced scattering which occurs on the long wavelength side of the chlorophyll absorption peaks. Gordon[6] first invoked anomolous dispersion to explain the enhanced reflectivity at 685 nm but later retracted this hypothesis in favor of chlorophyll fluorescence[7].

The one-dimensional model of radiative transfer has been effectively used by several researchers[1,8] to give fairly reliable reflectivities over a wide range of ocean parameters. It is the purpose of this paper to extend that model to account for chlorophyll fluorescence.

II. Theory

Let us consider a medium which contains a uniform distribution of particles which can scatter, absorb, and emit radiation. We will also assume that the radiation is restricted to either the upward (U) or downward (D) direction (see Fig. 1). Let $I_{\lambda'}^{U,D}(z)$ (w/m^2-nm) denote the spectral irradiance at wavelength λ' and depth z in the upward (U) or downward (D) direction in a medium which is infinitely deep. We will also adopt some of the notation used by Gordon[7] in what is to follow. It is easy to show that the net flux absorbed ,$F_A(z)$ (W/m^2), by a layer of thickness Δz for wavelengths in the spectral band of width $\Delta\lambda'$ is given by

$$F'_A(z) = \frac{d(I_{\lambda'}^U - I_{\lambda'}^D)}{dz}\Delta z \Delta\lambda' \qquad (1)$$

We will use the prime superscript to denote quantities which are functions of excitation wavelength. The number, $N_A(z)$, of photons absorbed per unit time per unit area is then given by

$$N'_A(z) = \frac{F'_A(z)\lambda'}{hc} \qquad (2)$$

where h is Planck's constant and c is the speed of light. The quantum efficiency $\eta(\lambda', \lambda_F)$ is defined to be the ratio of the rate at which photons are emitted

from Δz with bandwidth $\Delta\lambda_F$ to the rate at which they are absorbed by chlorophyll within bandwidth $\Delta\lambda'$. To find the fraction of photons absorbed by chlorophyll we must multiply eqn. (2) by $\beta_a^{\prime chl}/\beta_a^{\prime T}$ where $\beta_a^{\prime T}(m^{-1})$ is the total absorption coefficient and $\beta_a^{\prime chl}(m^{-1})$ is the chlorophyll absorption coefficient. Therefore the number of photons emitted per unit time per unit area is given by

$$N_F'(z) = \eta(\lambda', \lambda_F) \frac{\beta_a^{\prime chl}}{\beta_a^{\prime T}} N_A'(z) \tag{3}$$

Now let $J_F'(z)$ (w/m^2-m-nm) be the volume fluorescent flux at depth z in the medium, then

$$N_F'(z) = \frac{J_F'(z)\Delta z \Delta\lambda_F \lambda_F}{hc} \tag{4}$$

Combining eqns. (1), (2), (3) and (4) we get

$$J_F'(z)\Delta\lambda_F = \eta(\lambda', \lambda_F) \frac{\beta_a^{\prime chl}}{\beta_a^{\prime T}} \frac{d(I_{\lambda'}^U - I_{\lambda'}^D)}{dz} \frac{\lambda'\Delta\lambda'}{\lambda_F} \tag{5}$$

We will assume, as did Gordon[7], that the fluorescent emission band can be represented by a Gaussian spectral distribution centered at λ_{0F} = 685 nm with σ = 10.64 nm and therefore

$$J'_F(z) = n(\lambda',\lambda_F) \frac{\beta_a'^{chl}}{\beta_a'^{T}} \frac{d(I^U_{\lambda'} - I^D_{\lambda'})}{dz} \frac{\lambda' \Delta\lambda'}{\lambda_{0F}} \qquad (6)$$

$$\frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[- \frac{(\lambda_F - \lambda_{0F})^2}{2\sigma^2} \right]$$

As pointed out by Gordon[7], Forster and Livingston[9] have found experimentally that the quantum efficiency of chlorophyll a (in vitro) is independent of λ' for $\lambda' \leq \lambda_{0F}$ and falls rapidly beyond this value. We will therefore assume it to be a step function for $360 \leq \lambda' \leq 700$nm. Using eqn. (6) we will define

$$J_F(z) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[- \frac{(\lambda_F - \lambda_{0F})^2}{2\sigma^2} \right] \frac{n(\lambda_F)}{\lambda_{0F}} \qquad (7)$$

$$\int_{360}^{700} \frac{\beta_a'^{chl}}{\beta_a'^{T}} \frac{d(I^U_{\lambda'} - I^D_{\lambda'})}{dz} \lambda' d\lambda'$$

Now that we have obtained the source function we can proceed to the derivation of the differential equations governing the upward and downward fluorescent irradiance. It should be noted that the irradiance at the fluorescing wavelength due to scattered solar flux is separable from that due to fluorescing particles. Kattawar and Plass[10] already solved the one dimensional equation for scattered solar flux. We will make use of most of the notation in that article in the following derivation.

Referring to Fig. 1 let us consider adding an infinitesimal layer of thickness Δz to the bottom of a homogeneous medium of total thickness z. We will also assume that the fluorescence source function is symmetric in its emission i.e., equal amounts upward and downward. Let $r(z)$ and $t(z)$ denote the reflection and transmission operators respectively for a layer of thickness z. Let $H^U(z)$ and $H^D(z)$ denote the fluorescent irradiances emerging from the top and bottom of the layer respectively. Also let β_T denote the extinction coefficient at the fluorescing wavelength. Considering all processes up to first order in Δz we get

$$H^D(z+\Delta z) = H^D(z)(1-\beta_T\Delta z) + H^D(z)\; B\; r(z)\beta_T\Delta z + H^D(z)F\beta_T\Delta z + \frac{J_F(z)}{2}\,\Delta z\; r(z) + \frac{J_F(z)}{2}$$

where $F + B = \omega_0$ the single scattering albedo. It should be noted that we have neglected the self induced fluorescent term i.e., at 685 nm we have assumed that the excitation at this wavelength is caused solely by the external source and not the internal source. This assumption is clearly justified due to the fact that the fluorescing irradiance is small compared to the exciting irradiance. This equation has a sinple physical

interpretation (see Fig. 1). The first term on the right hand side of eqn. (8) represents the radiation emerging from the bottom of the layer of thickness z which passes through the sublayer Δz unscattered The second term represents the radiation emerging from z which backscatters in Δz and then is reflected from the overlying layer and then emerges from Δz unscattered. The third term is radiation emerging from z which forward scatters in Δz and emerges. The fourth term represents the fluorescent source emitting upwards which is reflected from the overlying layer and then emerges through Δz with no scatterings. The final term represents the downward fluorescing radiation from the layer Δz. Now passing to the limit as $\Delta z \to 0$ we get

$$\frac{dH^D(z)}{dz} = (1+r(z))\frac{J_F(z)}{2} - [(1-F) - r(z)B]\beta_T H^D(z) \quad (9)$$

Using the same notation we used for the one dimensional solution in reference 3 with the exception that λ has been changed to Ψ in this paper to avoid confusion with wavelength, we have

$$1-F=1-\omega_0+B=\alpha+B, \quad (10a)$$

$$r(z) = \frac{B[1-\exp(-\xi\beta_T z)],}{\Psi_2-\Psi_1\exp(-\xi\beta_T z)} \quad (10b)$$

$$t(z) = \xi \exp(-\xi\beta_T z/2)/[\Psi_2 - \Psi_1 \exp(-\xi\beta_T z)] \quad (10c)$$

where $$\alpha = 1-\omega_0, \quad (11a)$$

$$\xi = \Psi_2 - \Psi_1 = 2(\alpha^2 + 2\alpha B)^{1/2} \quad (11b)$$

$$\Psi_1 = \alpha + B - (\alpha^2 + 2\alpha B)^{1/2} \quad (11c)$$

$$\Psi_2 = \alpha + B + (\alpha^2 + 2\alpha B)^{1/2} \quad (11d)$$

To complete the solution to eqn. (9) we need to compute

$$\frac{d(I_{\lambda'}^U - I_{\lambda'}^D)}{dz} = -\frac{\xi'\beta_T'}{2}\left(\frac{B' - \Psi_2'}{\Psi_2'}\right)\exp(-\xi' z\beta_T'/2) \quad (12)$$

where the prime superscript means that we are considering a wavelength in the excitation process and the unprimed quantities denote their values at 685 nm. After performing some lengthy algebra, we can integrate eqn. (9) subject to the initial condition $H^D(0)=0$ and obtain

$$H^D(z) = \frac{K(\lambda_F)}{p} \int_{360}^{700} \delta' \left\{ \frac{(\Psi_2+B)}{\phi'_1} \exp\,(\phi'_1 z/2) + \frac{(\Psi_1+B)}{\phi'_2} \exp\,(-\phi'_2 z/2) - \frac{(\Psi_2+B)}{\phi'_1} - \frac{(\Psi_1+B)}{\phi'_2} \right\} \frac{\lambda'}{\lambda_{0F}} \frac{\beta'^{Chl}_a}{\beta'^{T}_a}\, d\lambda' \qquad (13)$$

where

$$\delta' \equiv \frac{(\Psi'_2 - B')\xi'\beta'_T}{4\Psi'_2}, \quad K(\lambda_F) = \frac{\eta(\lambda_F)}{\sqrt{2\pi}\,\sigma\lambda_{0F}} \exp\left[-\frac{(\lambda_F-\lambda_{0F})^2}{2\sigma^2}\right],$$

$$\phi'_1 = (\xi\beta_T - \xi'\beta'_T)/2, \quad \phi_2 = (\xi\beta_T + \xi'\beta'_T)/2$$

and

$$p = \Psi_2 \exp(\xi\beta_T z/2) - \Psi_1 \exp(-\xi\beta_T z/2)$$

It should be noted that $\lim_{z\to\infty} H^D(z) = 0$.

The differential equation for the upward irradiance, $H^U(z)$, exiting the top of the layer of thickness z is easily found to be

$$\frac{dH^U(z)}{dz} = t(z)[J_F(z)/2 + B\beta_T H^D(z)] \qquad (14)$$

Using eqns. (10c), (13), and (7) in eqn. (14) and performing the integration we obtain the following result subject to the initial condition $F^U(0) = 0$ and passing to the limit as $z \to \infty$.

$$H^U(\infty) = \frac{K(\lambda_F)\xi}{\Psi_2} \int_{360}^{700} \delta' \Big\{ \sum_{n=0}^{\infty} \frac{(\Psi_1/\Psi_2)^n}{\phi_2' + n\xi\beta_T} + \frac{B\beta_T}{\Psi_2} \sum_{n=0}^{\infty}$$

$$(n+1)(\Psi_1/\Psi_2)^n \Big[\frac{(\Psi_2+B)}{\phi_1'} \left(\frac{1}{-\phi_1' + (n+1)\xi\beta_T} - \frac{1}{(n+1)\xi\beta_T} \right)$$

$$+ \frac{(\Psi_1+B)}{\phi_2'} \left(\frac{1}{\phi_2' + (n+1)\xi\beta_T} - \frac{1}{(n+1)\xi\beta_T} \right) \Big\}$$

$$\times \frac{\lambda'}{\lambda_{oF}} \frac{\beta_a'^{chl}}{\beta_a'^{T}} H_o^D(\lambda')\, d\lambda' \qquad (15)$$

The factor $H_o^D(\lambda')$ appearing in eqn. (15) is the downward irradiance just above the ocean surface. It should be noted that in order to interchange the order of the integration and summation, which was used to evaluate the integrals over the variable z, the power series expansion must converge uniformly.

This was rather easy to establish for the integrand resulting from eqn. (14). We can reduce eqn. (15) to a rather simple form as follows. First for typical ocean models $\Psi_1/\Psi_2 \approx 10^{-6}$ and therefore we need only the $n=0$ term in the summation. Also since $(\beta\beta_T/\phi_1'\Psi_2) \ll 1$ we can also *neglect the second summation term*. We are therefore left with

$$H^U(\infty) \approx \frac{K(\lambda_F)\xi}{\lambda_{0F}\Psi_2} \int_{360}^{700} \frac{\delta'\beta_a'^{\,chl}\lambda' H_o^D(\lambda')}{\phi_2'\beta_a'^{\,T}}\, d\lambda' \tag{16}$$

Also over the entire spectral range $\beta' \ll \alpha'$ then

$$\Psi_2' \approx 2\alpha',\ \xi' \approx 2\alpha',\ \phi_2' \approx \alpha\beta_T + \alpha'\beta_T',$$
$$\delta' \approx \frac{\alpha'\beta_T'}{2}$$

Using these results in eqn. (16) and recognizing the fact that $\alpha\beta_T = (1-\omega_0)\ \beta_T = \beta_a^T$ we get

$$H^U(\infty) = \frac{K(\lambda_F)}{2\lambda_{0F}} \int_{360}^{700} \frac{\beta_a'^{\,chl}\lambda'}{(\beta_a^T+\beta_a^{T'})} H_o^D(\lambda')\,d\lambda' \tag{17}$$

which is a remarkably simple result.

III. Calculation

To test the results obtained we used the ocean model given in Table I. The chlorophyll a absorption coefficients were the in vivo values obtained from Morel and Prieur[1]. In Table II we present the results of our calculations for the quantum efficiency η for the same values of chlorophyll a concentration presented by Gordon[7]. We should note that the reflectivity R_s at 685 nm for zero chlorophyll concentration computed from the one dimensional model (see ref. 10) is B/Ψ_2 and for the model used in Table I give 0.095%. The extrapolated value shown by Gordon[7] using the data of Morel and Prieur[1] was 0.13%. The efficiency values in parenthesis in Table II used $R_s = 0.13\%$ whereas the first values used $R_s = 0.095\%$. In Table II we also present a comparison of the emission computed by using eqn. (15) the exact result, compared to the approximate equation (17). As can be seen the approximate result is accurate to less than 1% for the model presented. The range of the integral over wavelength was taken from 360 to 700 nm. The values presented are for the center of the emission line, namely 685 nm. The quantum efficiencies η obtained from this one-dimensional model are slightly lower than those obtained by Gordon using a quasi-single scattering approximation with full three dimensional transfer; however some of the difference could be due to the use of different

ocean models.

Using eqn. (18) an interesting upper bound can be placed on the quantity $R_\infty - R_S$. If we use a weighted mean for the quantum efficiency $\eta = 0.47\%$ then the upper bound will be $\approx 1.09\%$ and this result is only dependent on η, the spectral dependence of the chlorophyll a absorption coefficient, and downward irradiance.

IV. Conclusion

We have obtained an exact one-dimensional solution to the equations of transfer for chlorophyll fluorescence from an infinitely deep ocean with a uniform distribution of fluorescing bodies. We have also found that the general solution could be reduced to a very simple expression for the case where absorption dominates over scattering. This allowed us to establish an upper bound on the difference in reflectivities for a very high chlorophyll concentration. The results we obtained for the quantum efficiency are still low compared to the average value of 5% obtained by Kieffer[11]. Gordon also found this to be the case for the quasi-single scattering analysis. We agree with Gordon's conclusion that the problem may be in the spectral dependence of the in vivo absorption coefficient for chlorophyll a. The reason being that photon cellular absorption is not all converted to photon chlorophyll absorption. We also feel that

anomolous dispersion can <u>not</u> be ruled out considering the experimental evidence already presented for it. In a future paper we will use a simple dispersion model for the refractive index and starting from this calculate spectral scattering coefficients in the region of the chlorophyll absorption bands. This will introduce a concentration dependent scattering coefficient which may well provide enough <u>scattering</u> to produce the desired peak at 685 nm. The more crucial test is the <u>shape</u> of the reflectivity spectrum about the peak. The Gaussian structure gives added credence to the fluorescense hypothesis since anomolous dispersion scattering does not usually exhibit this structure.

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Table I. Scattering (β_S), absorption (β_a), and total extinction coefficients (β_T) for the water, hydrosol, and chlorophyll components of the ocean model as a function of wavelength. $H D_o$ is the downward irradiance just above the ocean surface.

λ (nm)	$H D_o$ (w/m^2-nm)		Water (m^{-1})	Hydrosol (m^{-1})	Chlorophyll a ($m^{-1}/(mg\text{-}m^3)$)
		β_a	3.80-2+	9.10-4	1.00-2
[illegible]50	0.826	β_S	1.20-2	3.00-2	-
		β_T	5.00-2	3.09-2	1.00-2
		β_a	2.20-2	8.50-4	1.20-2
[illegible]30	0.909	β_S	9.40-3	3.20-2	-
		β_T	3.14-2	3.28-2	1.20-2
[illegible]00	1.20	β_a	1.71-2	7.90-4	1.50-2
		β_S	7.60-3	4.00-2	-
		β_T	2.47-2	4.08-2	1.50-2
		β_a	1.53-2	7.30-4	2.00-2
420	1.51	β_S	6.10-3	8.60-2	-
		β_T	2.14-2	8.67-2	2.00-2
		β_a	1.45-2	6.90-4	2.35-2
[illegible]40	1.59	β_S	4.90-3	9.09-2	-
		β_T	1.94-2	9.16-2	2.35-2
		β_a	1.56-2	6.40-4	2.50-2
[illegible]50	1.84	β_S	4.10-3	8.70-2	-
		β_T	1.97-2	8.76-2	2.50-2
		β_a	1.76-2	6.10-4	2.25-2
[illegible]30	1.87	β_S	3.40-3	8.33-2	-
		β_T	2.10-2	8.39-2	2.25-2

+The notation -N means that the number is multiplied by 10^{-N}

Table 1 continued

λ(nm)	H^{D}_{o}(w/m2-nm)		water	hydrosol	chlorophyll a
		β_a	2.57-2	5.70-4	1.80-2
500	1.76	β_S	2.90-3	8.00-2	-
		β_T	2.86-2	8.06-2	1.80-2
		β_a	4.77-2	5.40-4	1.60-2
520	1.66	β_S	2.40-3	7.67-2	-
		β_T	5.01-2	7.72-2	1.60-2
		β_a	5.58-2	5.10-4	1.25-2
540	1.62	β_S	2.10-3	7.40-2	-
		β_T	5.79-2	7.45-2	1.25-2
		β_a	7.08-2	4.90-4	9.00-3
560	1.54	β_S	1.80-3	7.14-2	-
		β_T	7.26-2	7.19-2	9.00-3
		β_a	1.08-1	4.60-4	7.50-3
580	1.55	β_S	1.60-3	6.90-2	-
		β_T	1.10-1	6.95-2	7.50-3
		β_a	2.44-1	4.40-4	7.00-3
600	1.51	β_S	1.40-3	6.66-2	-
		β_T	2.45-1	6.70-2	7.50-3
		β_a	3.09-1	4.20-4	7.50-3
620	1.47	β_S	1.20-3	6.45-2	-
		β_T	2 45-1	6.70-2	7.50-3
		β_a	3.29-1	4.00-4	1.00-2
640	1.44	β_S	1.00-3	6.25-2	-
		β_T	3.30-1	6.29-2	1.00-2
		β_a	4.00-1	3.90-4	1.50-2
660	1.40	β_S	8.00-4	6.06-2	-
		β_T	4.01-1	6.10-2	1.50-2

Table I continued

λ(nm)	$H D_o$(w/m^2-nm)		water	hydrosol	chlorophyll a
680	1.36	β_a	4.50-1	3.70-4	1.90-2
		β_S	7.00-4	5.88-2	-
		β_T	4.51-1	5.92-2	1.90-2
700	1.31	β_a	6.50-1	3.60-4	2.00-3
		β_S	7.00-4	5.72-2	-
		β_T	6.51-1	5.76-2	2.00-3

Table II. Comparison of the exact emission using eqn. (15) with the approximate calculation using eqn. (17).

chl a(mg/m^3)	$R - R_s$	η (%)	% error
6.8	0.210	0.48 (0.53)	0.6
9.0	0.245	0.44 (0.48)	0.5
18.1	0.475	0.48 (0.52)	0.2

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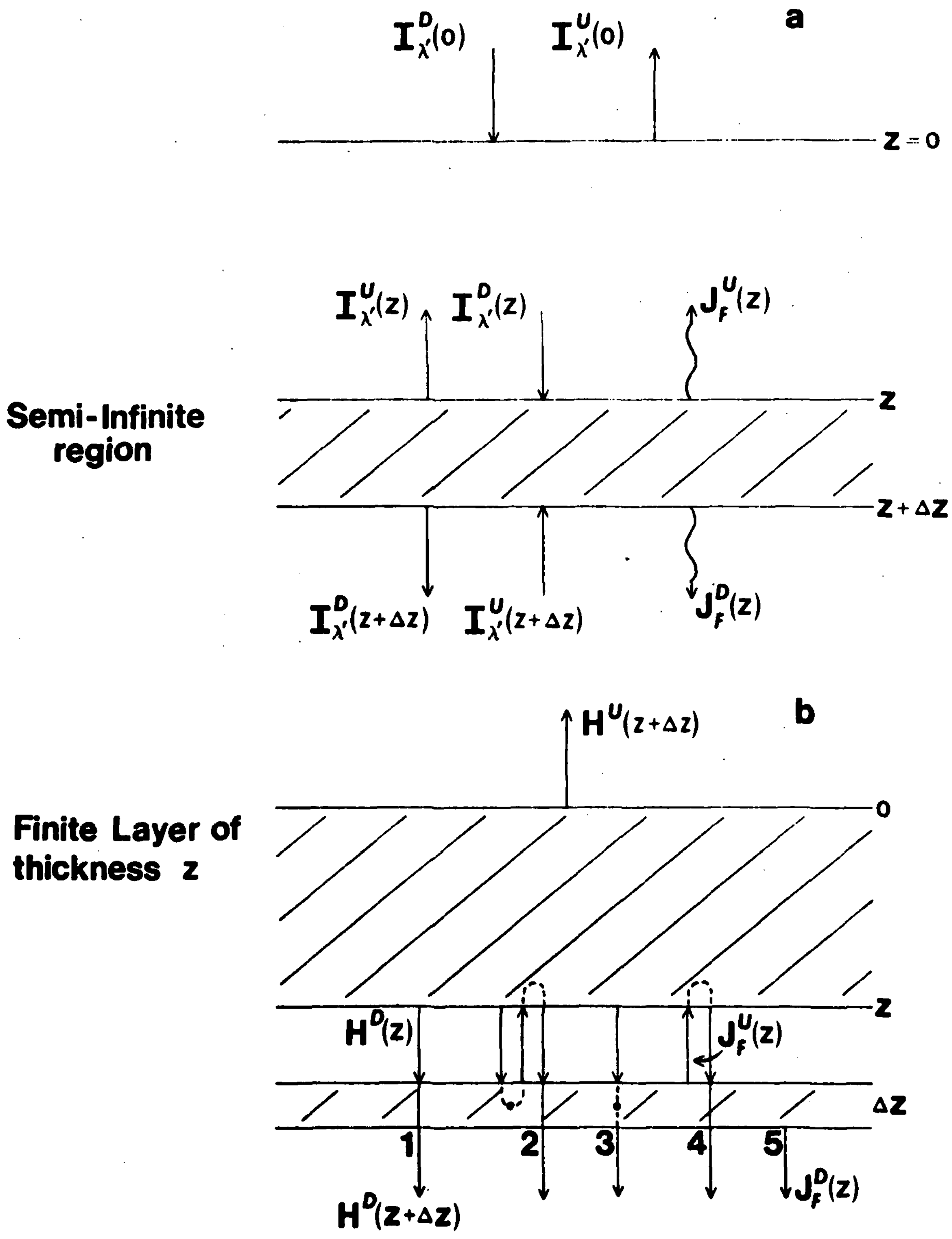
FIGURE CAPTIONS

Figure 1. (a) Schematic representation of source streams giving rise to fluorescent radiation. (b) Schematic representation of all first order processes encountered upon addition of an infinitesimal layer of thickness Δz to a layer of thickness z for the downward irradiance.


a
$I^{D}_{\lambda'}(0)$
$I^{U}_{\lambda'}(0)$
Z = 0
Semi-Infinite region
$I^{U}_{\lambda'}(z)$
$I^{D}_{\lambda'}(z)$
$J^{U}_{F}(z)$
z
z + Δz
$I^{D}_{\lambda'}(z+\Delta z)$
$I^{U}_{\lambda'}(z+\Delta z)$
$J^{D}_{F}(z)$
b
$H^{U}(z+\Delta z)$
Finite Layer of thickness z
0
$H^{D}(z)$
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$J^{D}_{F}(z)$
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Figure 1

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